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THE KINETICS OF LASER-PULSE VAPORIZATION OF URANIUM DIOXIDE BY MASS SPECTROMETRY

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The Kinetics of Laser Pulse Vaporization of Uranium Dioxide by Mass Spectrometry

Chuen-horng Tsai and Donald R. Olander

Safety analyses of nuclear reactors require knowledge of the evaporation behavior of UO_2 at temperatures well above the melting point of 3140 K. Currently used in the analyses are the extrapolations from the static measurements performed well below the temperature range of interest, based upon a few thermophysical or thermochemical models.

In this study, rapid transient heating of a small spot on a UO_2 specimen was accomplished by a laser pulse (see Fig. 1 for the set-up), which generates a surface temperature excursion. This in turn vaporizes the target surface and the gas expands into vacuum. The surface temperature transient was monitored by a fast-response automatic optical pyrometer. A computer program was developed to simulate the laser heating process and calculate the surface temperature evolution. A quadrupole mass spectrometer was used to identify and analyze the major vapor species in the vaporizing flow, and to measure the rate of evaporation from the target surface. The information yielded the partial vapor pressure of each species and the composition in the vapor jet.

The maximum surface temperatures investigated range from $\sim$3700 K to $\sim$4300 K. For the partial pressure of UO_2, the pressure-temperature relation $\log p(\mathrm{atm})=26.81-26089/T(\mathrm{K}) -5.594\log T(\mathrm{K})$ best fits to the experimental results. This p-T relation falls inside the confidence limits recommended in the literature assessment report[1] (see Fig. 2). The results of the vapor pressure and the vapor composition deduced from the experiments favor the Blackburn's model[2] for calculating the equilibrium partial pressure of each species compared to the other calculations in the literature.[3,4] It also suggests that the equilibrium vaporization, after taking into account the oxygen depletion on the surface,[5] successfully describes the vaporization process in the sub-millisecond transient. The measured time-of-arrival and the width of the mass spectrometric signals compared to the free molecular model (collisionless expansion) and hydrodynamic model (collision-dominated continuum flow) suggest that the free molecular expansion best represents the expansion process in vacuum. The fact that no dimers of any species were measured also suggests that collisions are negligible in the expansion into vacuum. The degree of ionization in the hot vapor was estimated from the mass spectrometer measurement of thermonic ions compared with the neutral molecules. The result was in good agreement with the calculation based on Sha's equation and effective "un-isolated" ionization potential.[6]

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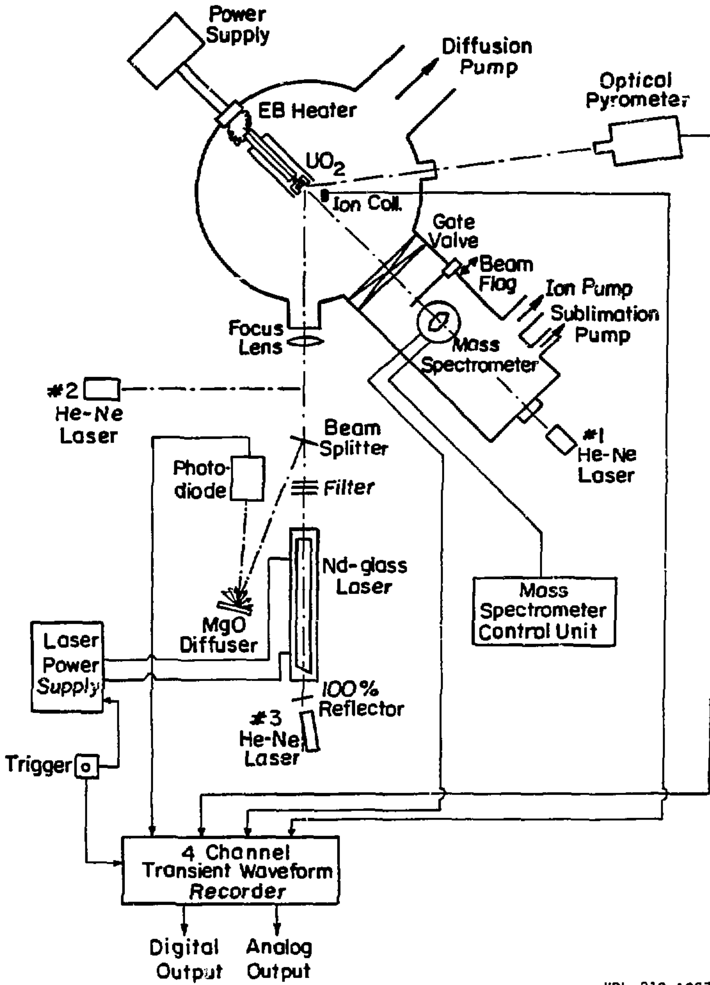

Fig. 1 The Apparatus Setup of the Laser-Induced Vaporization Study by Mass Spectrometry

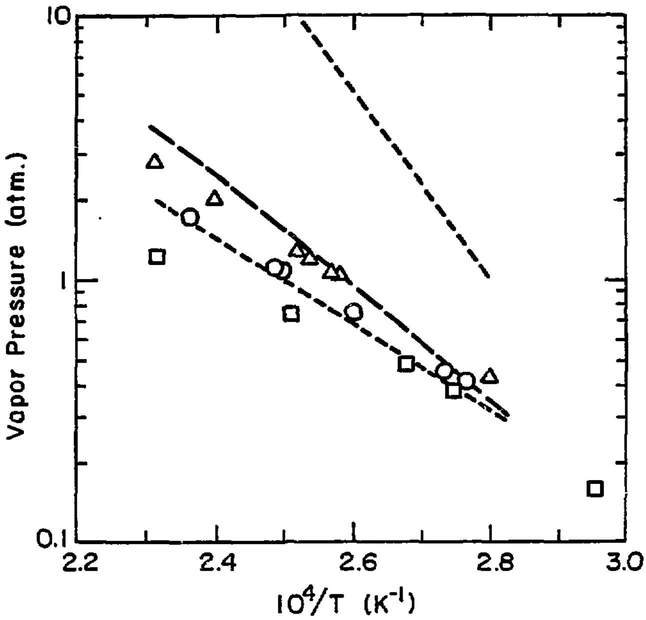

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Fig. 2 The Pressure-Temperature Relation for UO_2

— — — Blackburn's Model for UO_2 partial pressure[2]

------ recommended limits of total vapor pressure[1]

Δ This work fitted to $\log p = 26.81 - 26089/T - 5.594 \log T$ for UO_2 partial pressure (calc. temp.)

o This work fitted to $\log p = 24.22 - 24238/T - 5.033 \log T$ for UO_2 partial pressure (meas. temp.)

□ Mach disk Photographic measurements for total pressure (measured temp.)

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